

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

IN RE APPLICATION

OF:           WITTELER ET AL.

SERIAL No. 10/070,758

FILED:       JULY 11, 2003

FOR:         PROCESS FOR PREPARING POLYVINYLPYRROLIDONE-IODINE IN  
              AQUEOUS SOLUTION

Honorable Commissioner  
for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

D E C L A R A T I O N

I, Dr. Rainer Dobrawa, Dr. rer. nat., a citizen of the Federal Republic of Germany and residing at Stamitzstrasse 18, 68167 Mannheim, Germany, hereby declare as follows:

I am fully trained chemist, having studied at the University of Ulm, Germany, from 1995 to 2000, from which I received my Diploma in Chemistry;

From 2000 to 2004 I furthered my studies at the Institute of Organic Chemistry of the University of Würzburg, Germany, and I was awarded my doctor's degree by the said university in 2004;

I joined BASF Aktiengesellschaft of 67056 Ludwigshafen, Germany, in 2004, and have since been working in the field of polymer research, and am therefore fully conversant with the technical field to which the invention disclosed and claimed in application Serial No. 10/070,758 belongs.

I have studied the record of application Serial No. 10/070,758, and particularly the Office actions mailed on January 4, 2006 and

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August 24, 2006, and the prior art applied by the Examiner, in particular the teaching of *Denzinger et al.* (US 4,402,937).

It is my understanding that the Examiner still contends that the particular features of the process according to the claims of application Serial No. 10/070,758, are already known from *Denzinger et al.*.

I cannot share the Examiner's position as set forth in the Office actions of January 4, 2006 and August 25, 2006, for the following reasons. The teaching *Denzinger et al.* relates to a process for the preparation of PVP-iodine by reacting elemental iodine and PVP in aqueous solution. In col. 3 indicated lines 37 to 41 of US 4,402,937 it is stated that PVP having a K value from 8 to 50 can be employed. In col. 4 indicated lines 34 to 37 of US 4,402,937 it is stated that the PVP can be employed in a concentration from 10 to 60 % by weight.

The Examiner's position that *Denzinger et al.*'s disclosure allows for a combination of any of the K values and any of the PVP concentrations which fall within the ranges taught by *Denzinger et al.* is not deemed to be well taken. As I have pointed out in my first declaration, dated May 29, 2006, the viscosity of an aqueous solution of PVP strongly depends on the molecular weight of the PVP as reflected by the K-value, and on the concentration of PVP. This general background knowledge is reflected by the general teaching of *Denzinger et al.* in col. 4 indicated lines 34 to 37 where it is stated that the higher concentrations apply to PVP having a low K value, and vice versa. This general teaching is confirmed by the working examples of *Denzinger et al.* In example 3 a PVP having a K value of 12.5 is employed at concentration of 50 % by weight, while in examples 2, 4, 5a-c PVPs having K-values of 16.6 or 17 are employed at concentration of 40 % by weight and in example 1 a PVP having a K value of 31.5 is employed at a concentration of 30 % by weight.

The process according to the claims of application Serial No. 10/070,758 requires that an aqueous PVP solution is employed which has PVP concentration c that is larger than value resulting from the calculation  $100 \times [0.1 + 8:(K + 5)]$ . *Denzinger et al.* do not teach the utilization of PVP solutions which meet this requirement. In fact, each of the working examples of *Denzinger et al.* utilize PVP solutions having a PVP concentration that is lower than the value obtained from the calculation of claim 1 as I have shown in my first declaration.

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The inventors of application Serial No. 10/070,758 found out that the reaction taught by *Denzinger et al.* has several drawbacks in that mixing problems and iodine sublimation occur and practically insoluble sediments form as a result of mixing problems caused by the high viscosity of the PVP-solution. Moreover, the available iodine of the obtained PVP-iodine is not satisfactory (See my first declaration). Therefore, the reaction of *Denzinger et al.* is of limited applicability for the production of PVP-iodine.

Surprisingly, these drawbacks can be overcome by employing aqueous PVP solutions which fulfill the requirements of the calculation of claim 1 of application Serial No. 10/070,758.

It must be noted that the reaction of an aqueous PVP-solution with iodine leads to a multi-phase reaction mixture, because iodine is only sparingly soluble in water. Moreover the PVP leads to a high viscosity of the reaction mixture. The optimization of multi-phase reactions in a highly viscous reaction medium is very complex and in no case an optimization can be achieved by routine experimentation. Rather, much effort and tedious experimentation is required. In particular, it is impossible to predict the coaction of factors such as viscosity and mixing.

The inventors of application Serial No. 10/070,758 surprisingly found out that the calculation of claim 1 allows for a given PVP the finding of proper reaction conditions which overcome the drawbacks of the reaction taught by *Denzinger et al.*. According to the calculation of claim 1 of application Serial No. 10/070,758 an aqueous PVP solution has to be used, which has a concentration above a specific minimum concentration that depends on the K-value of the PVP used. According to the commonly accepted wisdom in the art a skilled person would have expected that decreasing the concentration of PVP would facilitate the reaction by reducing the viscosity of the reaction medium. Consequently, a skilled person would have performed the reaction taught by *Denzinger et al.* within the reaction parameters taught in the working examples of *Denzinger et al.* and he would not have used a given PVP at a concentration which is higher than the concentration given for said PVP in the working example. In complete contrast thereto, the invention disclosed in claim 1 of application Serial No. 10/070,758 requires to increase the concentration of PVP.

These findings are supported by the following additional comparative examples, which were run under my supervision. It must be

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noted that the following experiments were run in an equipment different from the equipment used in the experiments presented in my declaration of May 29, 2006. While being equipped with the same agitator, the reactors in my first declaration of May 29, 2006 and the reactors applied for the preparation of the examples presented in this declaration have different diameters as well as different height/diameter ratios, thus changing the stirring characteristics of the system. Therefore, the results presented in my earlier declaration cannot be compared with the results presented hereinafter.

The reaction of the aqueous PVP solution and iodine was performed in a 2-Liter glass reactor equipped with a crossbeam agitator at 400 rpm, a condenser and an internal temperature control. The reactor was heated by an external heating bath. If not stated otherwise, 330 g of PVP were charged as an aqueous solution into the reactor. The concentration is given in the following table. To this solution 3,72 g of formic acid were added. The solution was heated to 85°C internal temperature. Then 70,00 g of elemental iodine were added in 10 portions, with 5 min between each addition of iodine. After iodine addition was completed, the solution was stirred for 24 h at 85°C to ensure completion of the reaction.

For these experiments, the reaction was deemed to be completed when solid components could no longer be detected in the solution, which indicated that all solid iodine had been dissolved and had reacted. The solution was subsequently cooled to room temperature. The resulting PVP-iodine solution was decanted and the degree of formation of solid lumps in the reactor as well as incrustations in the condenser is assessed. Available iodine was determined by titration with thiosulfate. The results are summarized in the following table:

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Examp le	c <sup>1)</sup> [%bw]	K value	c <sub>min</sub> <sup>2)</sup> [%bw]	Obser- vations <sup>3)</sup>	Available Iodine [%] <sup>4)</sup>
4	35	30	32.9	+	7.0
Comp.4	28	30	32.9	-	5.8
5	60	12	57.1	↑	9.1
Comp.5	50	12	57.1	↓	8.8

- 1) PVP concentration of the utilized PVP solution in % by weight
- 2) minimum concentration calculated according  
$$c_{\min} = 100 \times [0.1 + 8:(K + 5)]$$
- 3) + no sedimentation, homogeneous mixing, no detectable solids  
- few solids, bad mixing, iodine sublimation  
↑ no iodine sublimation  
↓ iodine sublimation into the condenser
- 4) % by weight

The reactions according to examples 4 and 5 were good to handle, and neither sedimentation nor marked sublimation of iodine was observed. In particular the data of example 4 show for a PVP concentration slightly larger than the minimum concentration calculated by the relation in claim 1 of application Serial No. 10/070,758 the reaction is still acceptable. The comparison of example 4 with comparative example 4 shows that a decrease of the PVP concentration deteriorates the reaction performance, since mixing is bad and insoluble sediments are formed. Additionally, iodine sublimation in condenser occurs and the available iodine in the obtained product is decreased. Likewise, the comparison of example 5 with comparative example 5 shows that a decrease of the PVP concentration leads to strong sublimation of iodine, which might lead to a clogging of the apparatus, and to iodine loss. Moreover the available iodine is markedly decreased in the comparative example. Thus, the data clearly demonstrate the significance of utilizing a PVP solution having a PVP concentration above the calculated minimum concentration calculated according to the relation in claim 1 of application Serial No. 10/070,758.

I further declare that all statements made herein of my own knowledge are true and that all statements made on information or belief are believed to be true; and further that these statements are

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made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 101 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Signed at 67056 Ludwigshafen, this 17. day of January 2007.

  
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(Signature of Declarant)